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BOX PATENT APPLICATION

Assistant Commissioner for Patents
Washington, D.C. 20231

NEW APPLICATION TRANSMITTAL

Transmitted herewith for filing is the patent application of:

Inventor(s): Kevin L. Gering

For (title): PHOTOREACTOR WITH SELF-CONTAINED PHOTOCATALYST RECAPTURE

Enclosed are:

- ☒ 6 sheets of drawing(s). ☐ informal ☐ formal
- ☒ An assignment of the invention to Bechtel BWTX Idaho, LLC
- ☐ A certified copy of a _____ application.
- ☐ An associate power of attorney.
- ☒ Combined Declaration and Power of Attorney.
- ☒ Information Disclosure Statement.
- ☒ Form PTO-1449 (together with citations).

Fee Calculation (37 CFR 1.16)

CLAIMS AS FILED

| NUMBER FILED | NUMBER EXTRA | RATE | BASIC FEE \$710.00 |
|---|-----------------|-----------|-----------------------|
| Total Claims (37 CFR 1.16(c)) 20 -20= | 0 | X \$18.00 | = 0 |
| Independent Claims (37 CFR 1.16(b)) 4-3= | 1 | X \$80.00 | 80 |
| Multiple dependent claim(s), if any (37 CFR 1.16(d)) | | \$260.00 | = 0 |
| | | | TOTAL \$790.00 |

- ☒ Please charge Deposit Account No. 05-0565 in the amount of \$ 790.00.
- A duplicate copy of this sheet is attached.
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- ☒ The Commissioner is hereby authorized to charge the following additional fees by this paper and during the entire pendency of this application to Account No. 05-0565:
- ☒ Any additional filing fees required under 37 CFR 1.16(a), (f) or (g) (filing fees).
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- ☐ Any additional filing fees required under 37 CFR 1.16(e) (surcharge for filing the basic filing fee and/or declaration on a date later than the filing date of the application).
- ☐ Any patent application processing fees under 37 CFR 1.17.
- ☐ The issue fee set in 37 CFR 1.18 (at or before mailing of Notice of Allowance, pursuant to 37 CFR 1.311(b)).


 SIGNATURE OF ATTORNEY
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CCN 00-015273



BOX PATENT APPLICATION

Assistant Commissioner for Patents
Washington, D.C. 20231

LIT-PI-420, "PHOTOREACTOR WITH SELF-CONTAINED PHOTOCATALYST
RECAPTURE"

Dear Sir:

Enclosed is the subject patent application, six sheets of drawings, together with an executed Combined Declaration and Power of Attorney; a Patent Assignment, together with a Recordation Form Cover Sheet; an Information Disclosure Statement, together with Form 1449 and attachments; a fee sheet, in duplicate; and a return receipt postcard.

Sincerely,

Patricia A. Butikofer
Intellectual Property Administrator
Technology Transfer Office

PB

Enclosures

cc: R. J. Fisher, DOE-CH (w/o Encl)

Express Mail #EI568915460US

Atty. Dkt. No.: LIT-PI-420

**Title: PHOTOREACTOR WITH SELF-CONTAINED PHOTOCATALYST
RECAPTURE**

Inventors: Kevin L. Gering

1 **PHOTOREACTOR WITH SELF-CONTAINED**

2 **PHOTOCATALYST RECAPTURE**

3
4 **CONTRACTUAL ORIGIN OF THE INVENTION**

5 This invention was made with United States Government support under Contract
6 No. DE-AC07-94ID13223, now Contract No. DE-AC07-99ID13727 awarded by the
7 United States Department of Energy. The United States Government has certain rights in
8 the invention.

9
10 **BACKGROUND OF THE INVENTION**

11 **Field of the Invention**

12 This invention relates to systems for promoting photocatalytic reactions. More
13 particularly, the present invention relates to a reactor for causing photocatalytic reactions
14 in a liquid, and then recapturing the photocatalyst by exploiting the upward buoyancy of
15 the photocatalyst in relation to the liquid.

16 **State of the Art**

17 The value and importance of photocatalytic processes are well known, and
18 research into new and better photocatalysts and their uses is ongoing throughout the
19 world. There are many photocatalytic and photochemical mechanisms that have been
20 found to be beneficial in a wide range of applications. For example, through
21 photocatalysis, treatment of organic and inorganic contaminants in water is possible,
22 management of chemical reagents within a process can be improved, and many chemical

1 synthesis/manufacturing processes have been made possible which were either not
2 possible or practical before.

3 Although the ability to promote photocatalytic reactions is well established, the
4 *feasibility* of using these processes for practical water treatment, industrial processes, or
5 chemical synthesis has been elusive due to the challenges associated with processing and
6 handling finely dispersed semi-conductor photocatalyst materials in liquid. Use of such
7 photocatalyst suspensions is often impractical because of the economic constraints of
8 either sacrificing the photocatalyst (i.e. allowing disposal downstream) or recapturing it
9 in difficult filtration and recycling steps.

10 Heretofore, photoreactors based on photocatalyst suspensions have difficulties
11 providing both high oxidation efficiencies and acceptable processing flowrates while
12 achieving economical management of the photocatalyst. In addition, some water
13 treatment scenarios may mandate that the treated water be filtered prior to release to
14 remove suspended solids in order to satisfy turbidity and clarity criteria.

15 It would thus be desirable to have an efficient photoreactor which both prevents
16 the uneconomical loss of photocatalyst, while avoiding expensive and complicated
17 filtration systems.

18 In concert with a photoreactor, some method of reflecting and concentrating light,
19 typically sunlight, is generally desirable to promote higher efficiency in photochemical
20 reactions. Although solar technologies in general have proliferated into niche areas in
21 geographic regions where they are economically competitive, further research,
22 development, refinement, and growth of these technologies is needed to make them more

1 attractive and effective in locations that are not as amenable to solar technologies. One
2 area that could benefit from additional development as that of solar collector/reflector
3 devices. These devices act to capture solar energy through a large-surface mirrored area
4 and reflect it back to a target area much smaller than the mirror. The net effect of solar
5 collector and reflector devices is that solar energy may be concentrated far beyond the
6 natural incident solar radiation and directed at a specific target. The inventor has found
7 that there is a lack of such devices for vertically oriented targets (VOT's), such as
8 column-type reaction vessels, heat exchange pipes that depend upon thermosyphon
9 operation, etc. Most prior solar reflector devices, such as horizontally oriented trough
10 collectors, present limitations such as having a fixed reflective surface, and limited
11 adjustability and mobility.

12 It would therefore be desirable to have a solar collector/reflector which is mobile
13 and provides a reflective surface having an adjustable contour to accommodate the shape
14 and dimensions of the target, and which is also angularly adjustable to optimize its
15 position relative to the sun and the target.

16 17 **SUMMARY OF THE INVENTION**

18 It is therefore an advantage of the present invention to provide a photoreactor
19 which manages and recycles a photocatalyst through control of hydraulic and buoyant
20 forces.

1 It is another advantage of this invention to provide a photoreactor with a liquid-
2 phase photocatalyst suspension which does not require downstream auxilliary equipment
3 to recapture or filter out the photocatalyst.

4 It is another advantage of this invention to provide a photoreactor which allows
5 good quantum efficiencies, while reducing the mass of photocatalyst which is required to
6 promote photocatalytic reactions.

7 It is another advantage of this invention to provide a solar collector/reflector with
8 an adjustable contour reflective surface to accommodate the shape and dimensions of a
9 vertically oriented target.

10 The above and other advantages are realized in a system for the continuous use
11 and recapture of a catalyst in liquid, comprising: a reactor having a reaction zone with
12 generally downwardly flowing liquid, and a catalyst recovery chamber adjacent the
13 reaction zone containing a catalyst consisting of buoyant particles. The liquid in the
14 reaction zone flows downward at a rate which exceeds the speed of upward buoyant
15 migration of catalyst particles in the liquid, whereby catalyst particles drawn out of the
16 catalyst recovery chamber and introduced into the liquid in the reaction zone are drawn
17 downward with the liquid. A flotation chamber disposed below the reaction zone is
18 configured such that when the flowing liquid enters therein the liquid flow velocity drops,
19 allowing the buoyancy of the catalyst particles to cause them to flow back into the
20 catalyst recovery chamber, rather than being swept downstream.

21 In one illustrative embodiment, the photoreactor comprises a generally upright
22 outer cylinder; with an inner cylinder disposed inside and generally coaxially with the

1 outer cylinder, so as to define an annular reaction zone between the inner and outer
2 cylinders. The inner cylinder comprises inlets at its bottom for allowing inflow of
3 buoyant photocatalyst-coated microspheres, and an orifice disposed at its top for allowing
4 outflow of the photocatalyst microspheres into the downwardly flowing liquid in the
5 reaction zone. A flotation chamber is disposed at the bottom of the outer cylinder and is
6 configured to substantially reduce the flow velocity of the liquid upon exiting the reaction
7 zone, such that the buoyant microspheres are allowed to float upward through inlets of
8 the inner cylinder. The flotation chamber may also include a means for forming a vortex
9 in the liquid, such that the buoyant particles are further directed toward the inlets of the
10 inner cylinder. A photoreflector is provided for directing light into the reaction zone,
11 whereby the photocatalyst disposed on the microspheres induces desired chemical
12 reactions within the liquid while flowing through the reaction zone.

13 The invention provides an economical and practical photoreactor which is useful
14 for a wide variety of solar photocatalytic uses such as, for example, remediation of
15 ground and surface waters, production of hydrogen and oxygen from water, recycling of
16 costly chemical reagents such as chelating or complexing agents, as well as non-solar
17 photocatalytic processes which require specific wavelengths of light in concentrated form,
18 such as, for example, chemical processes driven by lower UV wavelengths, such as less
19 than 300 nm. Other advantages and features of the present invention will be apparent to
20 those skilled in the art, based on the following description, taken in combination with the
21 accompanying drawings.
22

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a cross-sectional view of the photoreactor according to the present invention;

FIG. 2A is a closeup side view of an alternative inlet cone configured for use with the photoreactor of FIG. 1;

FIG. 2B is a horizontal cross-sectional view of the alternative inlet cone of FIG. 2A;

FIG. 3 depicts a complete photocatalytic system configured for use with the photoreactor of the present invention;

FIG. 4 is a graph of performance data for the photoreactor during a trial run to treat cyanide-laden water;

FIG. 5 is a cross-sectional view of an alternative embodiment of the photoreactor of the present invention;

FIG. 6 is a closeup view of the novel vertically oriented adjustable reflector for use with the photoreactor of the present invention;

FIG. 7A is an elevation view of the solar reflector in relation to a VOT;

FIG. 7B is a plan view of the reflection pattern of the solar reflector about a VOT relative to incident solar radiation;

FIG. 7C is a plan view of variable positioning of the solar reflector about a VOT; and

FIG. 8 is a graph of the estimated effective solar energy received by the VOT during the trial run.

DETAILED DESCRIPTION OF THE INVENTION

Reference will now be made to the drawings in which the various elements of the present invention will be given numeral designations and in which the invention will be discussed so as to enable one skilled in the art to make and use the invention. It is to be understood that the following description is only exemplary of the principles of the present invention, and should not be viewed as narrowing the pending claims.

FIG. 1 is a cross-sectional view of a photoreactor 10 according to the present invention. The photoreactor 10 generally comprises an outer cylinder 12, an inner cylinder 14 disposed coaxially within the outer cylinder, an inlet chamber 16 at the top, and a flotation chamber 18 at the bottom. The annular space between the inner and outer cylinders forms a reaction zone 20 having a height H. The inner space of the inner cylinder 14 comprises a catalyst recovery chamber 32. At the top of the inner cylinder is a distributor cap 22 which forms an orifice 24 about the top of the inner cylinder which allows fluid communication between the catalyst recovery chamber 32 and the reaction zone 20. At the bottom of the inner cylinder is an inlet 26 which allows restricted fluid communication between the flotation chamber and the catalyst recovery chamber. The inlet 26 preferably comprises an inverted perforated cone, which will be described in more detail below.

The photoreactor 10 operates in a flow-down configuration, with the liquid stream (e.g. water) entering through a conduit 28 connected to the inlet chamber 16 in the top of the reactor, and flowing downward through the annular reaction zone 20. A photocatalyst solution comprising buoyant photocatalyst-coated beads dispersed in the liquid is drawn

1 out of the catalyst recovery chamber 32 in the inner cylinder 14 through the orifice 24 due
2 to a venturi/siphoning effect, and the catalyst beads are drawn down through the reaction
3 zone by the flow of the liquid. Upon reaching the bottom of the reaction zone, the flow
4 enters the flotation chamber, where, due to its larger dimension, the flow velocity slows,
5 allowing the buoyant force of the photocatalyst beads to cause them to float upward
6 through the inlet 26 of the inner cylinder 14, where the beads continue to float upward in
7 the catalyst recovery chamber 32 until they reach the distributor cap 22, ready to be
8 recycled into the reaction zone. Some concurrent upward fluid flow occurs to assist
9 catalyst recovery, based upon the outflow of fluid at the orifice 24 due to the above-
10 mentioned siphoning effect.

11 The reaction zone 20 is preferably about 1/8 inch thick, this thickness being
12 primarily dependent upon the opacity of the liquid and the need for light to pass through
13 the liquid to allow complete exposure. The thickness of the reaction zone also affects
14 hydraulic parameters, such as the downward velocity of the liquid and the related velocity
15 of the microspheres. The outer cylinder 12 of the photoreactor may be formed of any
16 suitably rigid and transparent material which is resistant to the chemicals to be
17 encountered in the reactor. For use of the reactor for the degradation of cyanide in water,
18 the inventor used a Type L Teflon FEP (fluorinated ethylene propylene) outer cylinder.
19 This material has excellent transparency to near-UV and visible light, and is chemically
20 resistant to most if not all of the liquid phase constituents expected in cyanide
21 contaminated water. It is also strong, rigid, and resistant to abrasion. The dimensions of
22 the outer cylinder will vary depending upon the application. The outer cylinder used by

1 the inventor was 3.5 inches in diameter, and configured such that the height H of the
2 reaction zone was approximately 32-34 inches. As discussed below, these dimensions
3 provided a suitably long residence time in the reaction zone to allow the desired
4 reactions.

5 The inner cylinder 14 is somewhat shorter than the outer cylinder 12, and has a
6 diameter approximately 1/4 inch less than the outer cylinder in order to create the 1/8"
7 annular reaction zone. The inner cylinder is made of rigid polystyrene that is preferably
8 provided with a reflective coating, such as Silverlux SA-85P reflective film made by 3M
9 Corporation. Materials other than polystyrene could be used so long as they are suitably
10 rigid and inert.

11 The flotation chamber 18 is preferably formed of a rigid, inert polymer. For the
12 trial run (discussed below) involving the photocatalytic degradation of cyanide in water,
13 the inventor used a flotation chamber made of clear polycarbonate material having a very
14 low transmittance (<2%) to UV wavelengths. This was done to restrict the incidence of
15 effective light, and thereby restrict photocatalytic reactions, to the reaction zone, to
16 thereby allow accurate appraisal of the effectiveness of the reactor itself. It will be
17 apparent that alternate design and dimensional specifications may be used for the
18 photoreactor to accommodate other applications. For example, beneficial variations
19 could be realized for the length and diameter of the reaction zone, the reaction
20 zone/annulus thickness, choice of constructions materials, etc.

21 The photocatalyst solution comprises an aqueous dispersion of hollow ceramic
22 microspheres which are coated with a photocatalyst, such as zinc oxide (ZnO), titanium

1 dioxide (TiO₂), or other semiconductor photocatalysts or photosensitive materials. The
2 photocatalyst used by the inventor for the degradation of cyanide in water was Degussa P-
3 25 TiO₂. It will be apparent that various photocatalysts may be selected for various
4 chemical applications. The substrate of the microspheres is type SLG hollow ceramic
5 microspheres made by PQ Corporation. The microspheres have diameters of from about
6 100µm to about 300µm. However, this preferred size may vary depending upon the
7 specific catalyst selected and the liquid and other chemical and physical factors involved
8 in the specific reactor setup.

9 The TiO₂ coating comprises about 20% to 30% of the weight of the coated
10 microspheres. The procedure for coating the ceramic microspheres with TiO₂ is known
11 by those skilled in the art, and is thoroughly discussed in Jackson, N. B. et al., *Attachment*
12 *of TiO₂ Powders to Hollow Glass Microbeads: Activity of the TiO₂-Coated Beads in the*
13 *Photoassisted Oxidation of Ethanol to Acetaldehyde*, Journal of the Electrochemical
14 Society, Vol. 138 No. 12 (1991). The coated microspheres are formed to have a specific
15 gravity of preferably about 0.7 to 0.8. It will be apparent that microspheres with a
16 different specific gravity may be selected depending upon the liquid to be introduced into
17 the reactor, the flow rate, etc.

18 As the liquid flows down through the reaction zone 20 under gravity flow at
19 approximately ambient temperature and pressure, the photocatalyst solution enters the top
20 of the reaction zone through the orifice 24 and mixes with the downward flowing liquid.
21 The photocatalyst solution is drawn through the orifice partly due to a venturi/siphoning
22 effect which is created at the lip 30 of the distributor cap 22. The mixture of liquid and

1 catalyst solution is carried downward due to the downward flow velocity of the liquid in
2 the reaction zone. The hydraulics of the photoreactor are specifically designed such that
3 the upward buoyant force of the microspheres in the solution is more than countered by
4 the downward flow of the liquid. While the photocatalyst is present and dispersed in the
5 liquid in the reaction zone, the light entering the reaction zone causes intended
6 photocatalytic reactions in the liquid. Fluid flow through the reaction zone also produces
7 mild turbulence among the microspheres, so as to minimize diffusion-limited reaction
8 rates.

9 Upon leaving the reaction zone 20, the mixture of liquid and photocatalyst
10 solution enters the flotation chamber 18. The flotation chamber is configured to
11 significantly reduce the downward flow velocity of the mixture, allowing the upward
12 buoyant force of the microspheres to overcome the downward flow velocity of the liquid.
13 In the photoreactor constructed and tested by the inventor, the flotation chamber had a
14 volume of approximately 10 liters. As the liquid slows, the microspheres begin to float
15 upward, toward the inlet 26 disposed at the bottom of the inner cylinder 14, allowing
16 passage into the catalyst recovery chamber 32 within the inner cylinder.

17 The inlet 26 is preferably a downwardly oriented perforated cone formed of high
18 density polyethylene (HDPE), with perforations large enough to admit the microspheres.
19 A closeup view of one embodiment of an inlet 34 is given in FIG. 2A. The inlet
20 generally comprises an inverted conical body 36 having a plurality of perforations 37
21 formed therein. The inlet 34 may also have radially extending vanes or fins 38 which
22 help direct the microspheres toward the openings in the cone, as described below.

1 Upon entering the catalyst recovery chamber 32 in the inner cylinder 14, the
2 microspheres continue their upward buoyant migration, dsignated by arrows 40 in FIG. 1,
3 until reaching the distributor cap 22 at the top of the catalyst recovery chamber, where
4 they are again siphoned out through the orifice 24, and into the liquid flowing
5 downwardly into the reaction zone 20.

6 The liquid in which the desired chemical reaction has occurred exits the flotataion
7 chamber 18 through outlet 42 and conduit 70, which are attached near the bottom of the
8 flotation chamber. It will be apparent that the flow rate of liquid out of the reactor should
9 be equal to the flow rate of liquid entering the reactor so as to prevent excessive
10 pressurization or depletion of the liquid within the system. The inventor has found that
11 the optimal range of liquid flow rates for the prototype system configured for detoxifying
12 cyanide in water was from approximately 1/4 to 2 gallons per minute. It will be apparent
13 that the hydraulic flow characteristics will need to be independently calculated for any
14 given system configuration and chemical combination to provide the desired flow rate,
15 velocity, etc.

16 It will also be apparent that it is desirable to assist the microspheres in migrating
17 toward the perforated cone. Mere upward force does not necessarily direct them to the
18 inlet 26. Accordingly, a moderate centripetal flow pattern or vortex, denoted by arrow
19 46, is produced within the flotation chamber 18 to increase the separation and recycle
20 efficiency for removing the microspheres from the bulk liquid phase. Because the
21 microspheres are buoyant (having specific gravities of approximately 0.7 to 0.8), they
22 tend to gravitate toward the center of the vortex-type flow pattern, as indicated by arrows

1 48, whereas the liquid (such as water, having a specific gravity of 1.0) migrates toward
2 the outside region of the vortex, as indicated by arrows 50. The inventor has found that
3 by forming a vortex around the perforated cone, the buoyant microspheres concentrate
4 toward the center of the flotation chamber 18. It will be apparent that the extent of the
5 vortex that is required will depend upon the dimensions of the flotation chamber, the
6 flowrate of the liquid through the system, the liquid density, and the population and size
7 distribution of the microspheres at the various specific gravities. A mild vortex pattern
8 (20 to 60 rpm) has been found to achieve the desired result for the range of process
9 flowrates, and may allow greater processing flowrates to be realized.

10 The desired vortex is produced by a large magnetic stirrer 52 disposed below the
11 flotation chamber. An optional or additional method of achieving the desired vorticular
12 flow pattern within the flotation chamber is to provide helical or spiral vanes 54 within
13 the annular reaction zone 20, which cause the downwardly flowing liquid to obtain a
14 circular or spinning flowing motion upon exiting the reaction zone. Hydraulic tests
15 performed by the inventor have shown that the magnetic stirrer used alone produced
16 satisfactory results, causing a vortex which contained a high percentage of the
17 microspheres.

18 FIG. 3 depicts a complete photocatalytic system 60 configured for use with the
19 photoreactor 10 of the present invention. The system generally comprises a reservoir 62,
20 a pump 64, the photoreactor 10, a solar collector/reflector 66 for directing light from the
21 sun 68 into the reaction zone, and tubing 70 for directing the fluid to the various parts of
22 the system. An outlet flow control valve 72 is provided at the outlet of the flotation

1 chamber 18, and a similar valve 74 is provided at the outlet of the reservoir. A sampling
2 valve 76 is also provided to allow fluid to be extracted from the system during operation
3 to test the efficiency of the system. A photocatalyst fill line 78 may also be connected to
4 the inlet chamber 16 to allow addition of photocatalyst at any time, whether before or
5 during operation of the photoreactor.

6 Fluid is moved through the system via the pump 64, which introduces liquid into
7 the inlet chamber 16 in the top of the reactor. From that point, fluid moves via gravity-
8 induced flow. The inventor has used a peristaltic pump for the pumping function, though
9 other pump types may be used. The prototype photocatalytic system 60 of FIG. 3 also
10 includes a temperature probe 79 disposed within the flotation chamber 18, for allowing
11 measurement of the temperature of the liquid as it flows through the system. It will be
12 apparent that the temperature probe may be placed at other locations in the system, and
13 that multiple temperature probes may be installed at different locations for more accurate
14 measurement, if desired.

15 A case study was performed using the photoreactor system of FIG. 3 with a
16 solution of mock wastewater containing $K_4Fe(CN)_6$, a highly stable cyanide-metal
17 complex, in water. The stability constant of this ferrous complex has been published at
18 values ranging from $10^{35.4}$ to 10^{47} . See Huiatt, J.L., et al., "Cyanide From Mineral
19 Processing", workshop proceedings, sponsored by National Science Foundation, and U.S.
20 Bureau of Mines (1983). In the case study, the concentration of the $Fe(CN)_6^{4-}$ ion was
21 such that there was a theoretical equivalency of 10 mg/L CN^- , assuming complete
22 dissociation of the cyanide-metal complex.

1 The mock wastewater had an initial volume of 30 liters, and a pH of 11.5 to 11.8.
2 Photocatalysis was promoted using a very low initial dose of 100 to 105 grams of
3 photocatalyst (approximately 0.13% TiO_2 by weight) for the overall mock wastewater
4 solution volume. An additional 60 grams of photocatalyst were added at $t=90$ min., this
5 addition being represented by the vertical dashed line 80 in FIG. 4. The operational flow
6 rate for the trial run was 1 gallon per minute (gpm) per reactor pass, which produced an
7 observed residence time of between 6 and 7 seconds for the microspheres moving
8 through the reaction zone. A net irradiation time of between 3 and 4 minutes was
9 accomplished for the trial run, this time interval representing the total elapsed time of
10 residence of the solution within the reaction zone.

11 Samples of the solution were taken during the test run and were analyzed for
12 cyanide (CN^-) and cyanate (CNO^-), an oxidized, stable form that is far less toxic than
13 cyanide. The analyses showed favorable destruction of the cyanide complex during the
14 trial run, followed by oxidation of a portion of the resultant aqueous free cyanide. FIG. 4
15 is a graph of performance data from the test run of the photoreactor. This graph shows
16 the relative levels of cyanide and cyanate levels during the test run. The initial
17 concentration of free cyanide (at time zero) was due to natural decomposition prior to the
18 run and photo-dissociation of easier or less stable CN-Fe ligands during the initial
19 preparatory period (approximately 15-20 min., beginning at the second vertical dashed
20 line 82 at the far left of FIG. 4) immediately preceding time zero, when the solution was
21 flowing through the system in open sunlight while the solar reflector 66 was covered.

1 FIG. 4 shows that there was a steady increase in the free cyanide (CN^-)
2 concentration (indicating good photo-dissociation of the cyanide-metal complex) and
3 there was a corresponding steady increase of cyanate (CNO^-) due to cyanide oxidation
4 through photocatalysis. The net cyanide (detected CN^- plus that which had been
5 oxidized) also increased throughout the run, reaching nearly 8 mg/L (i.e. 80% of the
6 possible 10 mg/L) by the end of the trial. In other words, the test run showed that the
7 photoreactor effectively destroyed approximately 80% of the cyanide complex through
8 photodissociation, followed by oxidation of a portion of the resultant aqueous free
9 cyanide.

10 The test run also indicated that the operational characteristics of the photoreactor
11 could be improved and refined, particularly from a hydraulic standpoint. For example, it
12 was noted that during hydraulic testing and during the test run that a small quantity of the
13 photocatalyst medium had flowed out of the flotation chamber 18 with the treated water,
14 and into the reservoir 62. The inventor believes that slight modifications to the position
15 and configuration of the perforated cone 36 should improve the efficiency of
16 photocatalyst recycling within the photoreactor 10. As noted above, it will be apparent
17 that alternate design, dimensional, and operational specifications may be used for the
18 photoreactor to accommodate other applications. For example, variations in the length
19 and diameter of the reaction zone, the annulus thickness, the flotation chamber
20 dimensions, choice of constructions materials, and operating parameters (e.g. liquid
21 flowrate and photocatalyst dosage) may be beneficial, depending upon the application.

FIG. 5 is a cross-sectional view of an alternative embodiment of the photoreactor of the present invention. In this embodiment, the photoreactor 100 is non-cylindrical, but instead is formed as a generally flat panel which is oriented in a non-vertical configuration, at some angle α relative to the horizon. The reactor comprises a reaction zone 102 and a catalyst recovery chamber 104, which are separated by a wall 106, which may have a reflective coating disposed thereon, similar to that disclosed above with respect to the inner cylinder 14 (FIG. 1). A layer of glass 108 or other suitable transparent material provides a top cover to the reaction zone, thus allowing adequate light of the appropriate wavelengths for the desired photochemical reactions in the reaction zone.

A distributor cap 110 is disposed at the top of the catalyst recovery chamber 104, and forms an orifice 112 at the top of the wall 106 to allow catalyst to flow out of the catalyst recovery chamber into the top of the reaction zone 102. A liquid delivery device, such as a pipe 114 with an outlet 116, delivers liquid to the top of the reaction zone. As the liquid flows down past the orifice, the catalyst solution is drawn out of the catalyst recovery chamber due to a slight venturi/siphoning effect. As described above, the flow rate of the liquid is selected such that the downward flow velocity exceeds the upward buoyant force of the microspheres.

One of the foremost issues of any photoreactor design is the degree of mixing or intimacy between the liquid and the photocatalyst particles. Because the reactor 100 is not vertical, the buoyant microspheres will tend to "crawl" along the inside surface of the reactor glass 108, as indicated by the exaggerated size microspheres 118 shown

1 congregated along a portion of the glass. This characteristic will tend to reduce the
2 intimacy of the liquid and the photocatalyst due to stagnation (or near stagnation) of the
3 particles against the outer wall of the reaction zone. Thus, while there may naturally be
4 some turbulence in the downwardly flowing liquid, which will tend to mix the
5 microspheres with the liquid, photochemical efficiency will tend to drop. One design
6 feature which can help reduce stagnation and promote mixing is to keep the reaction zone
7 thickness small, such as 2-3 mm, to exploit the buoyancy driven hydraulics.

8 To promote additional mixing, protuberances 120 may be formed on the inside
9 surface of the glass 108. Other obstacles may also be provided in the reaction zone 102
10 in order to help disrupt the flow of the liquid and promote mixing. Finally, to further
11 reduce migration problems with the photocatalyst particles, the angle α should preferably
12 be maintained at from about 75° to 90° relative to the horizon. The system could be
13 configured such that the angle α is adjustable, and varies to follow the sun for optimal
14 solar exposure. However, it will be apparent that any change in α will tend to have a
15 profound effect on the hydraulic characteristics of the system. Accordingly, it is
16 considered undesirable to make α variable.

17 Beyond the lower end of the reaction zone 102 is a flotation chamber 122, which
18 has a large size such that the velocity of the flowing liquid drops rapidly upon entering
19 the flotation chamber. One of the benefits of the embodiment of FIG. 5 is that the
20 creation of a centrifugal flow field is not needed within the flotation chamber. The lower
21 portion 124 of the reaction zone curves downwardly, such that the lowest extremity 126
22 of the wall 106 is approximately vertical, or curves slightly past vertical, for any given

1 angle α . With this configuration, when the liquid enters the flotation chamber and slows
2 down, the upward buoyant force of the microspheres will cause them to begin to migrate
3 upward toward the inlet 128 of the catalyst recovery chamber 104, as indicated by arrows
4 130. Because the lower end 126 of the wall 106 is vertical or past vertical, this upward
5 tendency will not direct the microspheres back toward the lower end of the reaction zone.
6 Meanwhile, the liquid will gradually flow toward the outlet 132 of the flotation chamber,
7 as indicated by arrows 134. It will be apparent that the flotation chamber must be of a
8 size such that the liquid slows sufficiently to allow the catalyst migrate upwardly, such
9 that catalyst will not be swept downstream into the outlet 132.

10 The configuration of FIG. 5 provides several benefits. It is simple, and may not
11 require a solar reflector, depending largely upon the selected angle α . Rather, the reactor
12 may directly absorb incident solar radiation 136 for the desired photochemical reaction.
13 However, concentrated solar energy could also be supplied using a series of conventional
14 flat-plate reflectors. This configuration is believed not to be as desirable or efficient as
15 the vertically oriented reactor configuration of FIGs. 1 and 3, unless the entire
16 reflector/reactor system were configured to rotate to follow the sun, or a system of
17 mirrors were employed. Nevertheless, the reactor provides for positive recapture of
18 photocatalyst without the need for complicated filtration systems which tend to reduce
19 the operating flow rate of the system as a whole.

20 As depicted in FIG. 3, and shown more particularly in FIG. 6, the inventor has
21 developed a vertically oriented variable contour solar collector/reflector 66 for use with
22 the photoreactor of the present invention. The reflector 66 comprises a mosaic reflective

1 surface 200 which provides true 3-dimensional adjustability. It can be adjusted in
2 contour, tilt, and overall position to accommodate virtually any vertically oriented target
3 (VOT) surface, regardless of its size and dimensions. As used herein, the term vertically
4 oriented target (VOT) refers to an object having a height greater than its width. The VOT
5 may be cylindrical, like the photoreactor of FIG. 1, or it may have any other shape or
6 cross-section. Advantageously, the reflector 66 is mobile, compact, and can be used with
7 solar or artificial light.

8 The reflector surface 200 comprises a plurality of generally vertical reflective
9 plexiglass pieces 202 which are coated with a reflective film which has a high reflectance
10 to light sources that are known to promote photo-catalytic reactions (e.g. solar energy in
11 the UV and lower visible wavelengths). A preferred reflective film which is suitable for
12 photocatalysis is Silverlux SA-85P made by 3M Corporation. Other reflective films may
13 be more suitable for other applications of the reflector. For example, some applications
14 may call for certain discrete wavelengths of light. Reflective films which reflect only
15 certain wavelengths of light are available for such applications.

16 The individual reflector pieces 202 are preferably 2x24 inches x 3/8 inch thick
17 coated plexiglass, laid side-to-side on a mobile, adjustable, rigid frame 204, which can be
18 manipulated to yield various contours. The frame 204 generally comprises two side
19 pieces 206, having rear vertical supports 208 and front vertical supports 209 extending
20 upwardly therefrom on each side, which are pivotally interconnected by transverse rods
21 218. The transverse rods are preferably 3/4 in. diameter metal pipe, and cross bracing,

1 such as cables or rope lines 220, is provided between the side pieces 206 to increase the
2 strength of the frame.

3 It will be apparent that the size and number of individual reflector pieces will
4 depend upon the size of the VOT and the intensity of reflected light desired. As shown,
5 the reflector 200 comprises two adjustable contour surfaces, an upper surface 210 and a
6 lower surface 212, which are supported on the frame. The frame also includes casters
7 214 to make it mobile, and by virtue of its mobility, the entire device can be rotated
8 axially around a VOT as shown in FIG. 7C, to maximize the amount of concentrated light
9 reflected onto the VOT. As discussed below, the inventor has found that this reflector
10 design has a theoretical maximum concentration factor of 10 for the photoreactor
11 described above.

12 The maximum total number of reflector pieces 202 which are used will depend
13 upon how many can be positioned on the framework for a given framework
14 configuration. In the test run discussed above, the inventor used a reflector with 74
15 pieces, having a net reflective surface area of approximately 24 ft². It will be apparent
16 that any number of reflector pieces may be used, depending upon the size of the VOT and
17 the particular application. The width of the reflector pieces may vary, but should be
18 selected such that the width of each reflector piece is less than the diameter or side
19 dimension (in the case of a non-cylindrical VOT) of the VOT.

20 The reflector pieces 202 are connected to the framework 204 with a hook and loop
21 type fastener system (e.g. Velcro®). This system comprises two nylon straps 216
22 (preferably 1.5 in. wide and 12 ft. long), one for each of the upper and lower contour

1 surfaces, 210 and 212, the straps being covered with hook and loop material and extended
2 transversely across the frame between the vertical supports 208. One strap supports the
3 upper edge of the upper contour surface, and the other strap supports the lower edge of
4 the lower contour surface. Corresponding hook and loop material (not visible) is
5 connected to each reflector piece, to hold the pieces to the straps in a manner well known.

6 The lower edge of the upper contour surface and the upper edge of the lower
7 contour surface are each supported by Velcro covered flexible rods 217. These rods are
8 preferably formed of fiberglass that is 1/4" wide and 8 ft. long, and provide a flexible yet
9 rigid arc-like structure to support the contours. The rods 217 are attached to the front
10 vertical support 209 with bolts, and are supported at midspan by the top transverse rod
11 218 which includes Velcro material where the flexible rod contacts the transverse rod.

12 The length of the nylon straps 216 can be adjusted to achieve a variety of contours
13 for the contour surface 200, ranging from a deep semi-circular curve to a nearly flat
14 contour. To do this, the framework side pieces 206 are pivoted about the ends of the rods
15 218 by selectively lengthening the front tension member 222 and shortening the rear
16 tension member 224, or vice versa. This action either draws the ends of the nylon straps
17 216 and flexible rods 217 closer together, or extends them farther apart. By pivoting the
18 frame and adjusting the length of the straps and flexible rods, a user can adjust both the
19 contour and the net effective reflective surface area of the reflector. At the same time, the
20 relative vertical angle of the reflective surface 200 will also change since the relative
21 length of nylon straps 216 to the span between left and right front supports 209 will
22 dictate the vertical orientation of the reflector pieces 202. The greatest reflective surface

1 area is achieved by setting the framework pivot in a wide position, with tension member
2 222 long, and tension member 224 short.

3 The advantages of the reflector configuration and its adjustability are clearly
4 shown in FIGs. 7A-7C. FIG. 7A shows that the upper and lower reflective surfaces, 210
5 and 212, respectively, are disposed at different vertical angles relative to the sun 70 due
6 to their varying vertical position with respect to the VOT. FIG. 7B shows a plan view of
7 the collector/reflector 66 located in its horizontal position with respect to the VOT. By
8 virtue of the gravitational pull on the straps 216 and flexible rods 217, the curved
9 reflective surface 200 naturally assumes a semi-elliptical form, which reflects solar
10 radiation onto the sides of the VOT oriented away from the sun. It will be apparent that
11 as the sun moves across the sky during any given day, the position of the
12 collector/reflector 66 must also change for optimal exposure. FIG. 7C shows the
13 collector/reflector in position A when the sun 220 is in position A, and in position B
14 when the sun is in position B.

15 In the preferred embodiment, a pyranometer 226 with a light sensor (e.g. LiCor
16 LI-250 Light Meter) may be attached to some part of the frame 204 to allow a user to
17 measure the solar flux reaching the reflective surfaces. This allows a user to optimally
18 orient and configure the reflector 66 for maximum solar efficiency for any given
19 situation. The net effective solar energy received by a VOT can be estimated by knowing
20 the incoming (ambient) solar flux, the reflectance of the reflective surface, a light
21 concentration factor (usually estimated), and the surface area of the VOT being impinged
22 with concentrated light. As noted above, the inventor has calculated a theoretical

1 maximum concentration factor f_c of 10 for the configuration used in the trial run. The
2 concentration factor is defined as the ratio of the net reflective surface area to the VOT
3 surface area being impinged by concentrated light. In practice, the realized value of f_c
4 will be less than the theoretical maximum value, and will depend largely on the available
5 reflective surface area, the distance between the reflective area and the VOT, the shape
6 and dimensions of the VOT, and the precision with which the reflector elements are
7 aligned with the VOT.

8 A graph of the estimated effective solar energy received by the VOT during the
9 trial run is given in FIG. 8. From this graph it can be seen that the solar reflector yielded
10 more than a three-fold increase in the amount of solar energy at the VOT surface. The
11 deviant data point designated 230 was apparently the result of temporarily higher solar
12 reflectance from a nearby building roof during the course of the trial run. During the trial
13 run it was noted that the theoretical maximum f_c was not achieved, although concentrated
14 solar light ($f \approx 3$ to 4 as observed) was delivered to the photoreactor. This was largely
15 due to the reflector requiring frequent manual adjustments to track the position and angle
16 of the sun in relation to the VOT.

17 It will be apparent that automatic solar tracking and adjustment systems could be
18 incorporated into the reflector system, whereby optimal solar orientation may be
19 maintained continuously as the sun's vertical and horizontal angles change. Likewise, an
20 intelligent control system could be incorporated to provide optimal adjustment of the
21 contour of the reflective surface to maximize the amount of concentrated solar light
22 received by the VOT. For example, a controller receiving feedback from one or more

1 pyranometers and/or light sensors could be configured to control motors or other
2 actuators for automatically pivoting the frame 204 to shorten or lengthen the straps 216,
3 thereby automatically adjusting the shape of the reflector surfaces to maximize reflected
4 light. Other improvements will be apparent to those skilled in the art, particularly
5 through adopting well known advances which have been made in the field of solar
6 collectors, control systems, etc.

7 The reflector 66 advantageously allows for true 3-dimensional adjustability of the
8 reflective arrays by allowing adjustment of the tilt of the reflector elements, adjustment of
9 the geometry of the support frame, and axial movement of the entire solar array around a
10 VOT. The reflector system thus makes the capture and use of solar energy more
11 practical, and thus more economical, for targets that have previously been viewed as
12 unconventional or non-amenable to solar applications. It also makes the use of solar
13 energy practical in regions where it may otherwise be considered uneconomical. In
14 addition to its use with the disclosed photoreactor and other similar photo-chemical
15 applications, it may also be used for photo-thermal applications (e.g. water heating), as
16 well as photo-optical uses (e.g. providing concentrated solar light to optic fiber bundles
17 for home, commercial, and industrial lighting).

18 In sum, the invention described herein provides an economical and practical
19 photoreactor which is useful for a wide variety of photocatalytic uses for environmental
20 and industrial applications, as well as photocatalytic precesses which require specific
21 wavelengths of light in concentrated form. Due to its unique hydraulic design, the
22 photocatalyst is managed and recycled within the reactor while providing favorable

1 operating conditions for photo-catalytic processes, thus reducing the cost per gallon for
2 the desired reactions. Additionally, through careful selection of construction materials
3 and design criteria, the photoreactor and associated solar concentrator/reflector maximize
4 the amount of light energy that impinges the reaction zone within the photoreactor.

5 The inventor has specifically investigated the use of the photoreactor of the
6 present invention for treatment of water to degrade cyanide contamination therein. Other
7 advantageous uses of the invention include both environmental and industrial
8 applications. For example, the invention may be used for remediation of ground and
9 surface waters to remove a variety of organic and inorganic pollutants, rather than prior
10 methods which typically rely upon activated carbon or chemical oxidation processes.

11 The invention may also replace prior electrolytic methods for the production of hydrogen
12 and/or oxygen from water, as is desirable in the transportation and utilities industries for
13 alternate energy sources and power generation. In the electroplating and manufacturing
14 industries, the invention is useful for reuse and destruction of cyanide, recycling of costly
15 chemical reagents such as chelating or complexing agents, for water treatment, and so
16 forth. The photoreactor would replace prior systems which relied upon pH manipulation,
17 chemical oxidation, chelation, liquid-liquid extraction, chemical precipitation,
18 electrowinning, etc. The photoreactor may also be used in a variety of industries to
19 replace various liquid phase processes requiring chemical reagents (e.g. oxidizers) for
20 chemical synthesis. Such an application could bypass some chemical pathways for
21 production of specialty chemicals, thus decreasing sidestream wastes. Other useful
22 applications will be apparent to those skilled in the art.

1 It is to be understood that the above-described arrangements are only illustrative
2 of the application of the principles of the present invention. Numerous modifications and
3 alternative arrangements may be devised by those skilled in the art without departing
4 from the spirit and scope of the present invention and the appended claims are intended to
5 cover such modifications and arrangements.

I CLAIM:

1 1. A system for the continuous use and recapture of a catalyst in liquid,
2 comprising:
3 an inlet;
4 an upwardly oriented reaction zone coupled to the inlet and including a generally
5 downwardly flowing carrier liquid;
6 a flotation chamber disposed below and communicating with the reaction zone,
7 configured such that the downward flow velocity of the liquid is reduced
8 upon entering the flotation chamber to a velocity wherein the buoyancy of
9 the catalyst particles causes them to flow upward;
10 an outlet coupled to the flotation chamber; and
11 an upwardly oriented catalyst recovery chamber coupled above the flotation
12 chamber and containing a catalyst consisting of buoyant particles
13 suspended in the liquid, the catalyst recovery chamber, flotation chamber,
14 and reaction zone configured such that catalyst particles are drawn out of
15 the catalyst recovery chamber into the reaction zone, and are drawn
16 downward with the liquid into the flotation chamber for recycling to the
17 catalyst recovery chamber.

1 2. The system of claim 1, wherein the reaction zone is substantially vertical.

1 3. The system of claim 2, further comprising:
2 a catalyst recovery inlet disposed at a bottom of the catalyst recovery chamber and
3 approximately centrally located within the flotation chamber; and
4 rotational flow structure coupled to the flotation chamber for inducing rotational
5 flow about the center of the flotation chamber, such that the buoyant catalyst particles are
6 urged toward the catalyst recovery inlet to the catalyst recovery chamber.

1 4. The system of claim 3, wherein the catalyst recovery inlet comprises a
2 downwardly oriented cone having openings formed therein.

1 5. The system of claim 4, wherein the catalyst recovery inlet further
2 comprises fins disposed on an outer side of the cone, the fins configured to assist catalyst
3 particles in entering the openings formed in the cone.

1 6. The system of claim 3, wherein the rotational flow structure comprises a
2 magnetic stirrer disposed adjacent the flotation chamber.

1 7. The system of claim 1, wherein the reaction zone is substantially non-
2 vertical.

1 8. A photoreactor system, comprising:
2 a generally upright outer cylinder;

3 an inner cylinder disposed generally coaxially within the outer cylinder and
4 defining an annular reaction zone therebetween, the inner cylinder having an inlet at a
5 bottom thereof for allowing inflow of buoyant photocatalyst particles, and an orifice
6 disposed at a top thereof for allowing outflow of the buoyant photocatalyst particles into
7 downwardly flowing liquid in the reaction zone;

8 means for directing light into the reaction zone, whereby the photocatalyst
9 particles may induce photo-chemical reactions within the liquid; and

10 a flotation chamber communicating with and disposed at a bottom of the outer
11 cylinder and configured to substantially reduce the flow velocity of the liquid upon
12 exiting the annular reaction zone to a velocity wherein the buoyant microspheres are
13 allowed to float upward through the inlet of the inner cylinder.

1 9. The photoreactor of claim 8, further comprising means for inducing
2 rotational flow in the flotation chamber about the center of the flotation chamber, such
3 that the buoyant catalyst particles are urged toward the inlet to the catalyst recovery
4 chamber.

1 10. The photoreactor of claim 9, wherein the means for inducing rotational
2 flow is selected from the group comprising a magnetic stirrer disposed adjacent the
3 flotation chamber and spiral vanes disposed within the annular reaction zone.

1 11. The photoreactor of claim 8, wherein the photocatalyst is selected from the
2 group consisting of zinc oxide (ZnO) and titanium dioxide (TiO₂).

1 12. The photoreactor system of claim 8 wherein:
2 the liquid comprises cyanide contaminated water;
3 the inlet to the inner cylinder comprises a downwardly oriented perforated cone
4 approximately centrally located within the flotation chamber;
5 the means for directing light into the reaction zone comprises a variable contour
6 reflector;
7 and further comprising:
8 a water inlet for introducing the liquid into the top of the photoreactor;
9 means for inducing rotational flow around the perforated cone such that the
10 buoyant catalyst particles are urged toward the cone; and
11 a water outlet at a bottom of the flotation chamber for removing treated water.

1 13. The system of claim 12, wherein the photocatalyst comprises buoyant
2 ceramic beads coated with titanium dioxide.

1 14. The system of claim 13, wherein the coated beads are from approximately
2 100 µm to 300 µm in diameter.

1 15. The system of claim 12, wherein the variable contour reflector comprises:
2 a movable frame having two side portions;
3 a central connecting member pivotally connecting the side portions of the frame;
4 at least two flexible straps attached between the side portions of the frame, the
5 location of attachment being spaced from the central connecting member; and
6 a plurality of substantially vertical reflecting members disposed upon the straps
7 having their reflective surfaces generally commonly oriented, forming a curved mosaic
8 reflective surface;
9 whereby a user may adjust the contour of the reflective surface by pivoting the
10 side portions of the frame so as to adjust the curvature of the straps.

1 16. A method of recapturing photocatalyst particles in a liquid, comprising the
2 steps of:

- 3 (a) flowing a liquid downwardly through a reaction zone;
4 (b) introducing buoyant photocatalyst particles into the downwardly flowing
5 liquid, such that the photocatalyst particles are drawn downward through the reaction
6 zone with the flowing liquid; and
7 (c) recycling the particles through a catalyst recovery chamber by reducing
8 the downward flow velocity of the liquid beyond a bottom of the reaction zone, such that
9 the buoyant photocatalyst particles are allowed to float upward through the catalyst
10 recovery chamber to the reaction zone for reintroduction into the downwardly flowing
11 liquid.

1

17. The method of claim 16, further comprising the step of:

2

(d) directing light into the reaction zone such the photocatalyst particles may

3

promote chemical reactions within the liquid;

1

18. The method of claim 16, wherein the step of reducing the downward flow

2

velocity of the liquid beyond the reaction zone further comprises the step of discharging

3

the liquid into a flotation chamber below the reaction zone, the flotation chamber having

4

dimensions such that the velocity of the flowing liquid is substantially reduced from the

5

downward velocity of the liquid in the reaction zone.

1

19. The method of claim 18, further comprising the step of:

2

(e) inducing rotational flow of the liquid about the center of the flotation

3

chamber, such that the buoyant photocatalyst particles are urged toward an inlet to a

4

catalyst recovery chamber approximately centrally disposed within the flotation chamber.

1

20. The method of claim 18, further comprising the step of:

2

(f) withdrawing the liquid from the flotation chamber.

ABSTRACT

1 A system for the continuous use and recapture of a catalyst in liquid, comprising:
2 a generally vertical reactor having a reaction zone with generally downwardly flowing
3 liquid, and a catalyst recovery chamber adjacent the reaction zone containing a catalyst
4 consisting of buoyant particles. The liquid in the reaction zone flows downward at a rate
5 which exceeds the speed of upward buoyant migration of catalyst particles in the liquid,
6 whereby catalyst particles introduced into the liquid in the reaction zone are drawn
7 downward with the liquid. A slow flow velocity flotation chamber disposed below the
8 reaction zone is configured to recapture the catalyst particles and allow them to float back
9 into the catalyst recovery chamber for recycling into the reaction zone, rather than being
10 swept downstream. A novel 3-dimensionally adjustable solar reflector directs light into
11 the reaction zone to induce desired photocatalytic reactions within the liquid in the
12 reaction zone.

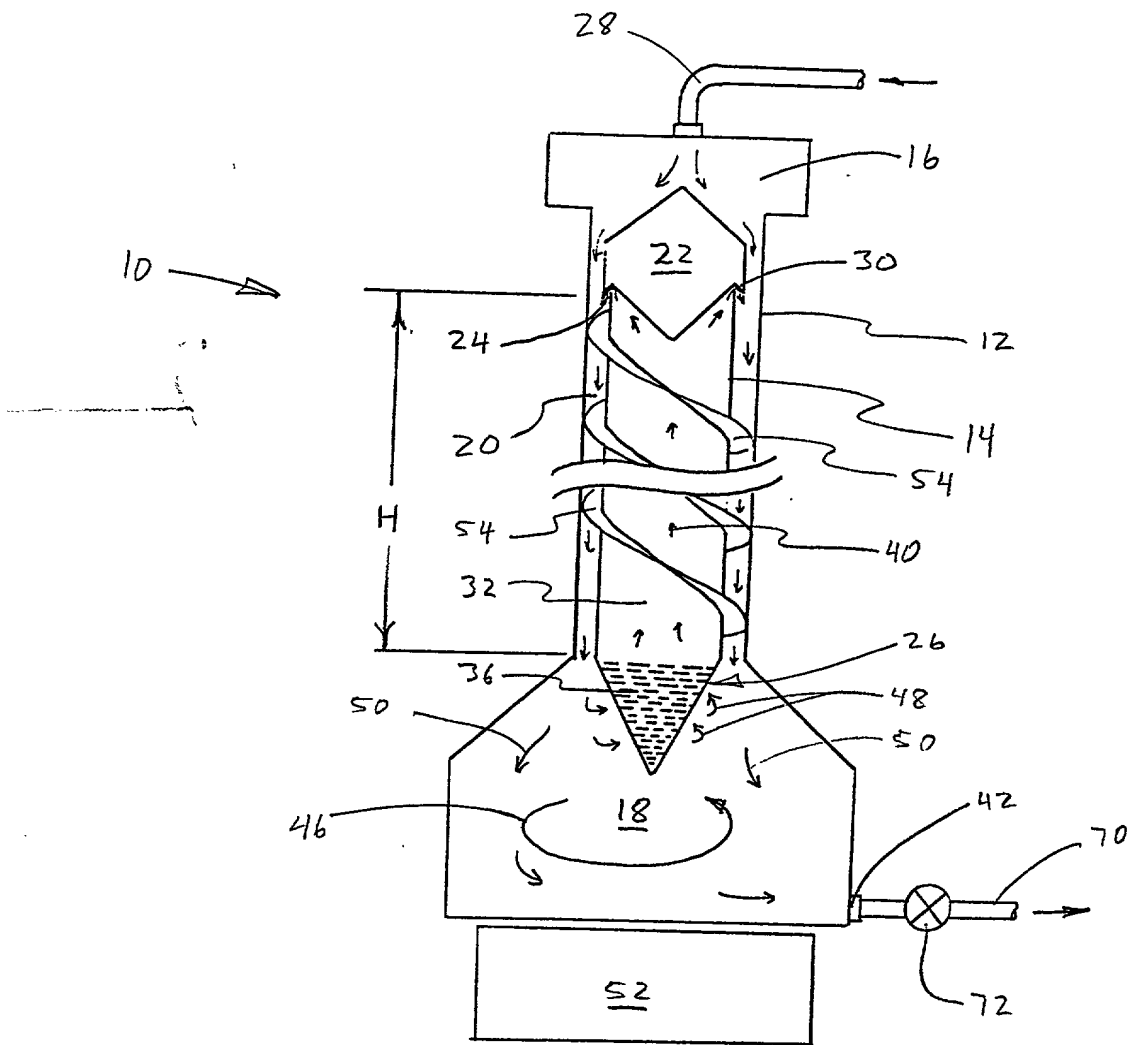


FIG. 1

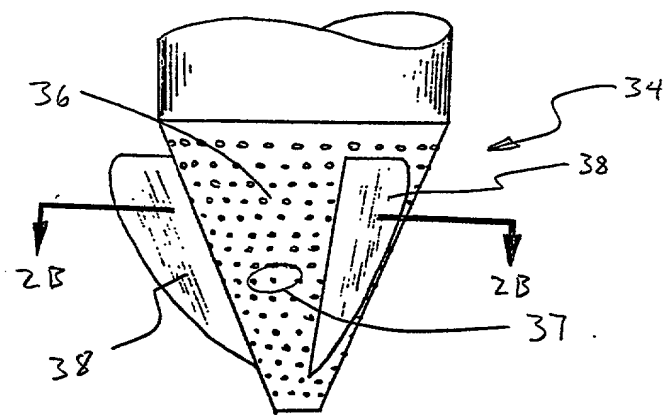


FIG. 2A

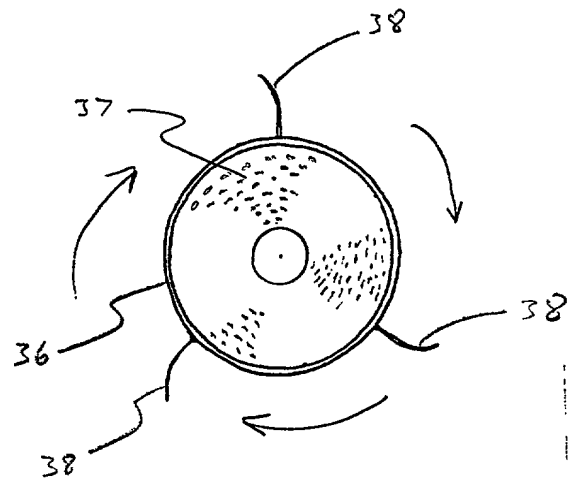


FIG. 2B

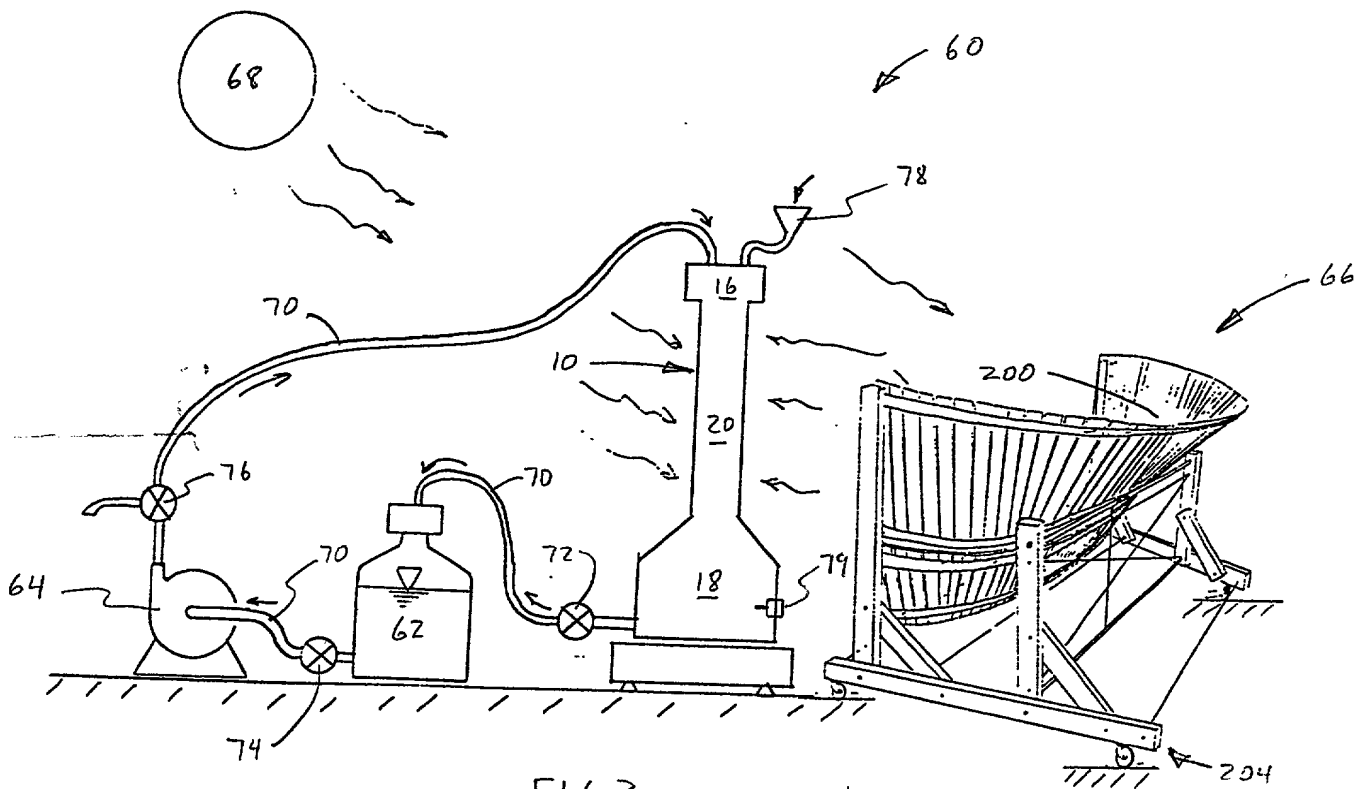


FIG. 3

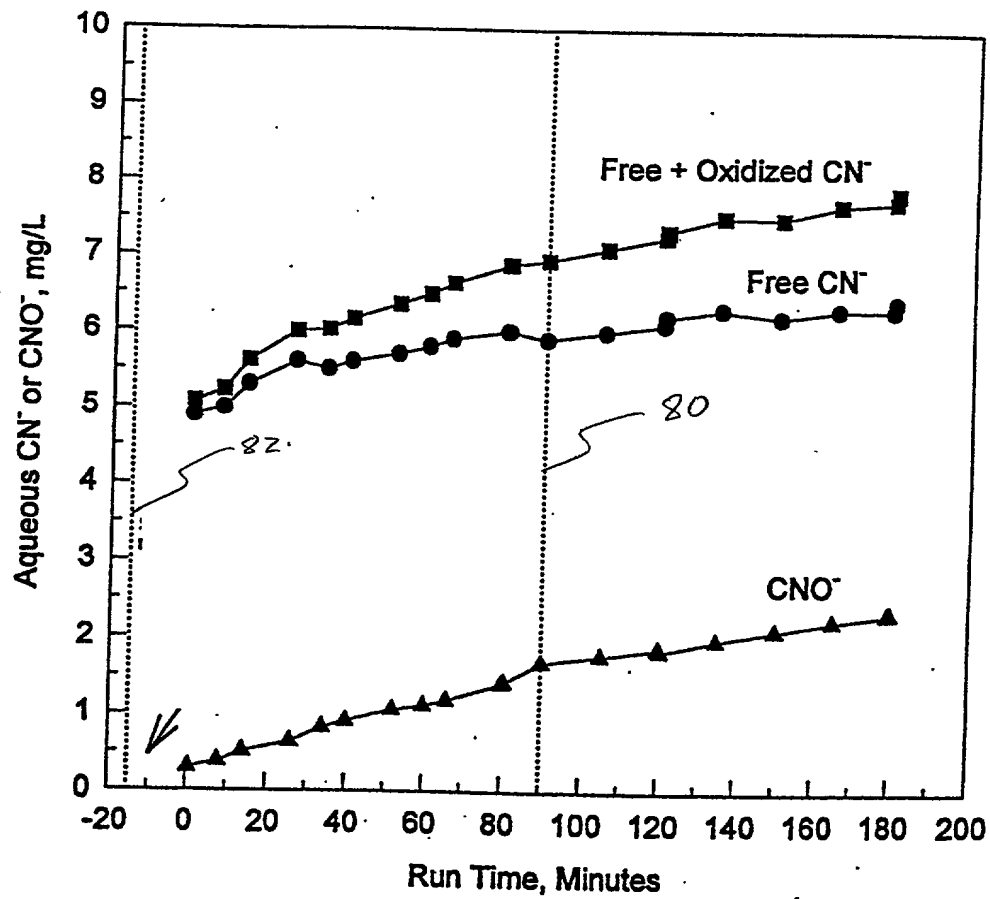


FIG. 4

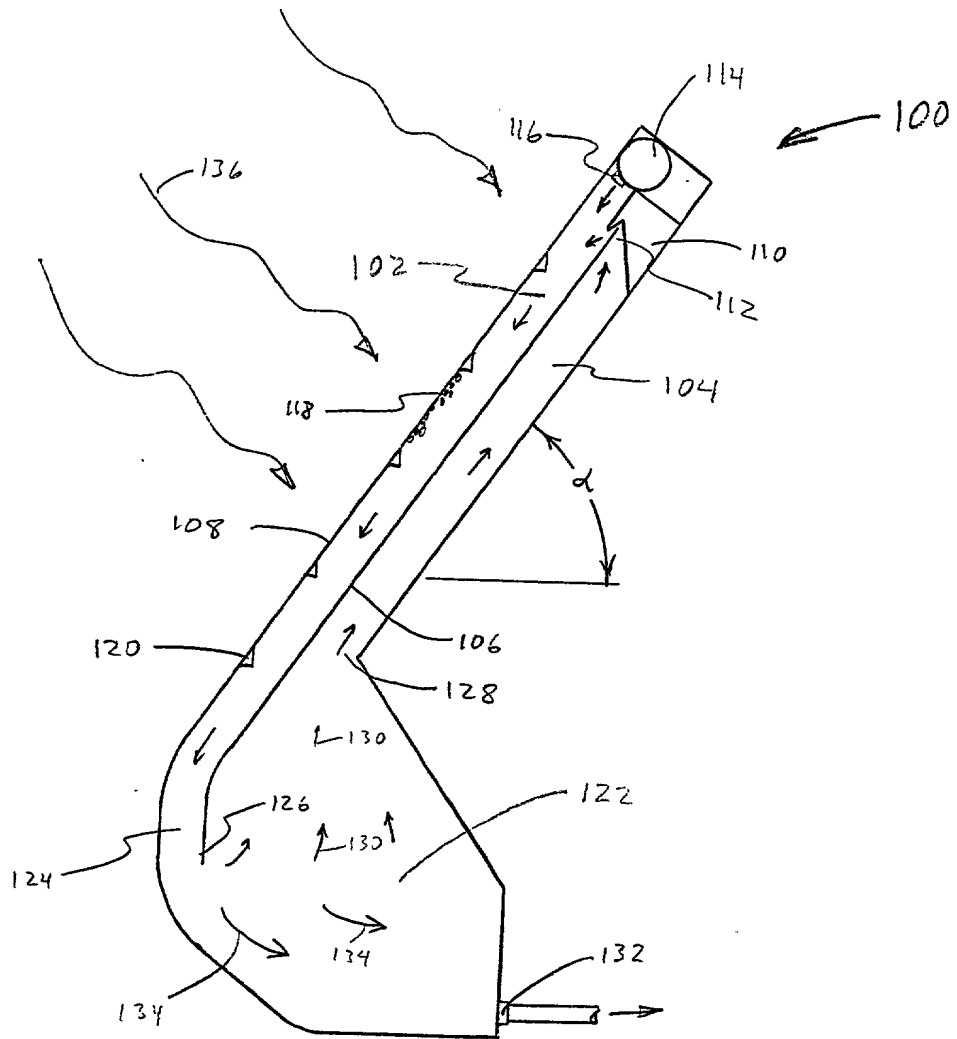


FIG. 5

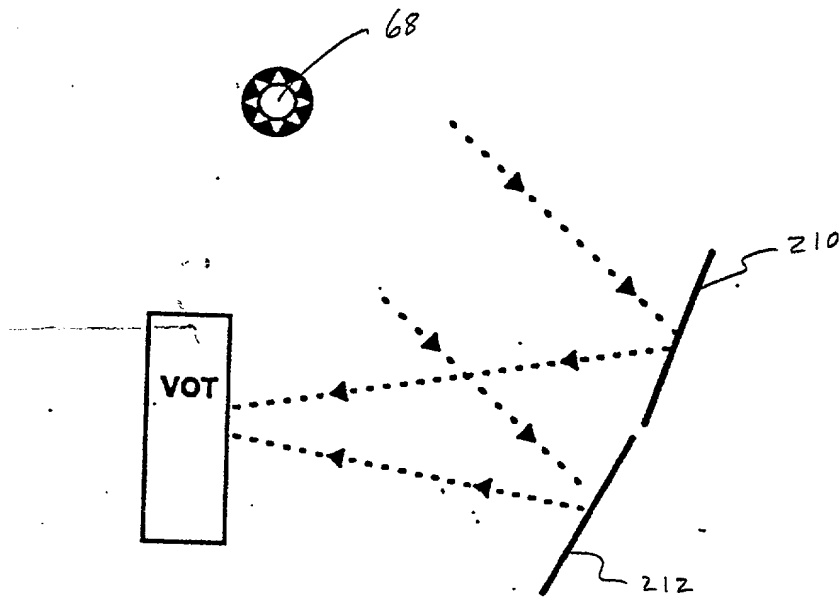


FIG. 7A

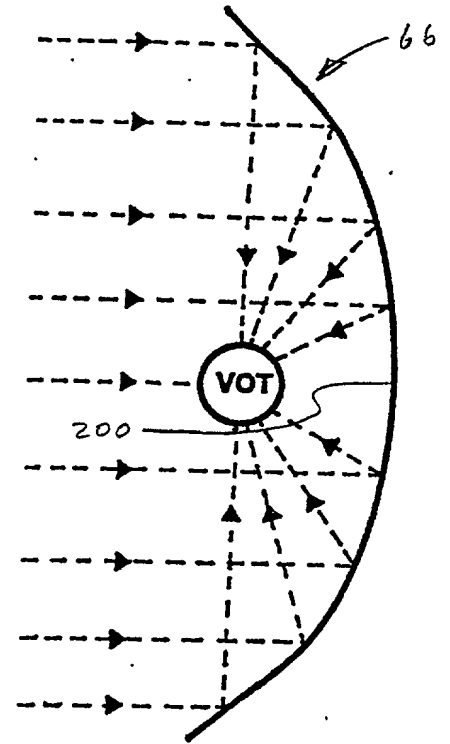


FIG. 7B

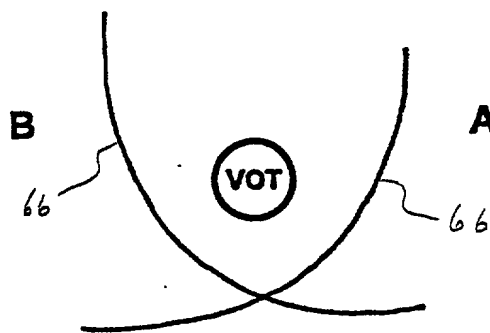
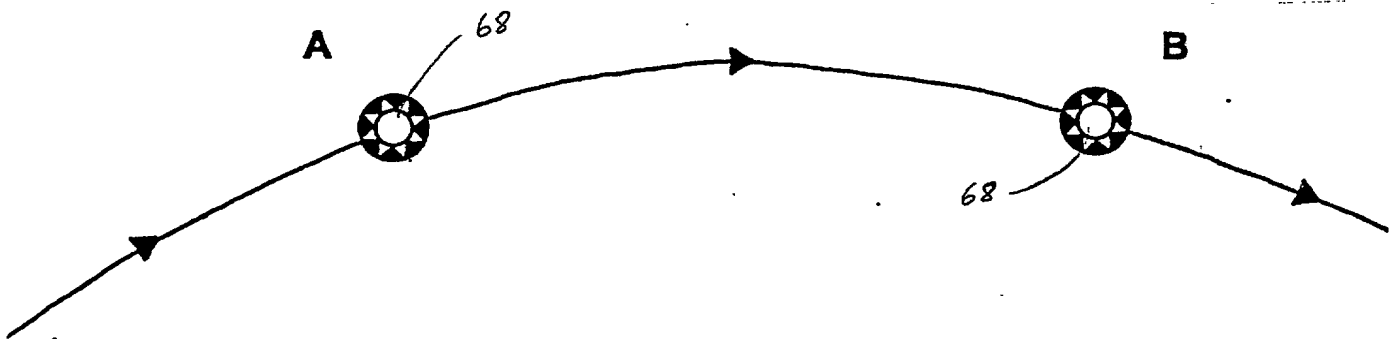


FIG. 7C

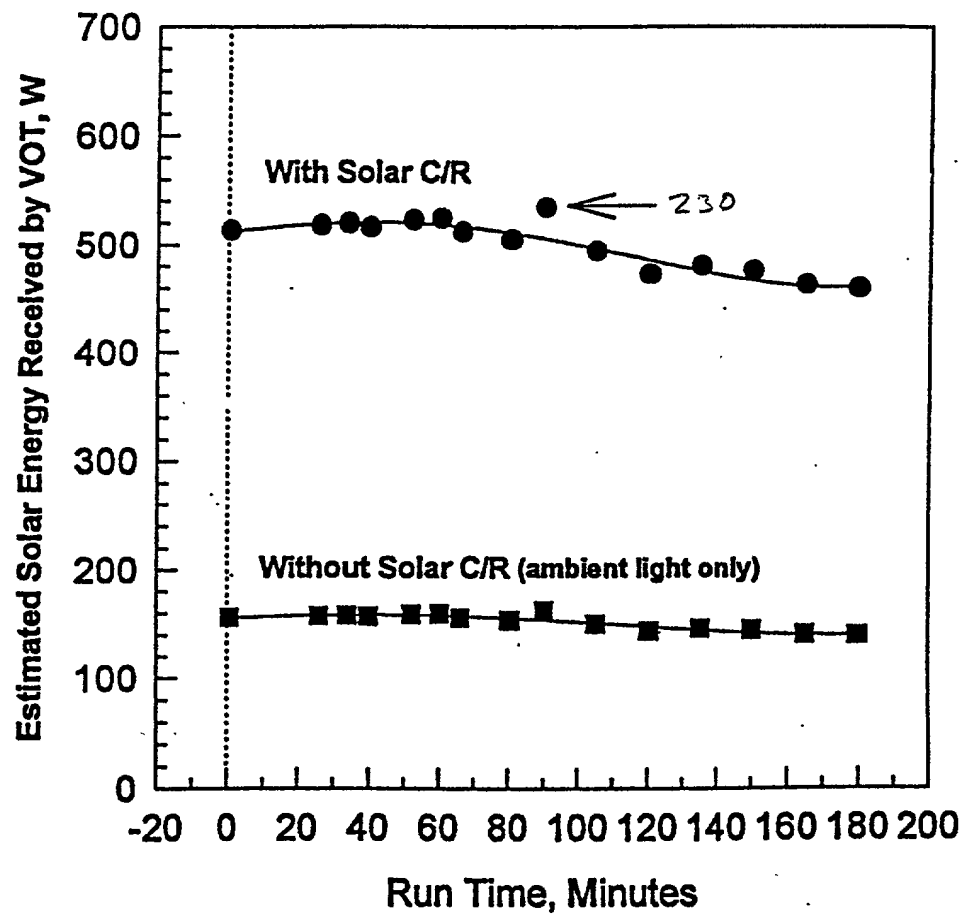


FIG. 8

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| | | |
|---|-------------------------------|-----------------|
| DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION (37 CFR 1.63) <input checked="" type="checkbox"/> Declaration Submitted with Initial Filing OR <input type="checkbox"/> Declaration Submitted after Initial Filing (surcharge (37 CFR 1.16 (e)) required) | Attorney Docket Number | LIT-PI-420 |
| | First Named Inventor | Kevin L. Gering |
| | COMPLETE IF KNOWN | |
| | Application Number | / |
| | Filing Date | |
| | Group Art Unit | |
| | Examiner Name | |

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Photoreactor with Self-Contained Photocatalyst Recapture

the specification of which (Title of the Invention)

☒ is attached hereto
OR

☐ was filed on (MM/DD/YYYY) as United States Application Number or PCT International

Application Number and was amended on (MM/DD/YYYY) (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

| Prior Foreign Application Number(s) | Country | Foreign Filing Date (MM/DD/YYYY) | Priority Not Claimed | Certified Copy Attached? | |
|-------------------------------------|---------|----------------------------------|--------------------------|--------------------------|--------------------------|
| | | | <input type="checkbox"/> | YES | NO |
| | | | <input type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> |
| | | | <input type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> |
| | | | <input type="checkbox"/> | <input type="checkbox"/> | <input type="checkbox"/> |

☐ Additional foreign application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto:

I hereby claim the benefit under 35 U.S.C. 119(e) of any United States provisional application(s) listed below.

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|-----------------------|--------------------------|--|
| | | <input type="checkbox"/> Additional provisional application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto. |

[Page 1 of 2]

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DECLARATION — Utility or Design Patent Application

I hereby claim the benefit under 35 U.S.C. 120 of any United States application(s), or 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

| U.S. Parent Application or PCT Parent Number | Parent Filing Date (MM/DD/YYYY) | Parent Patent Number (if applicable) |
|--|---------------------------------|--------------------------------------|
| | | |

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

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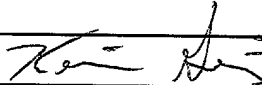
| Name | Registration Number | Name | Registration Number |
|-----------------|---------------------|------|---------------------|
| W. Gary Goodson | 22,387 | | |

☐ Additional registered practitioner(s) named on supplemental Registered Practitioner Information sheet PTO/SB/02C attached hereto.

Direct all correspondence to: ☐ Customer Number or Bar Code Label ☒ Correspondence address below

| | | | | | |
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| Country | US | Telephone | 208-526-9469 | Fax | 208-526-8339 |

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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| Name of Sole or First Inventor: | | <input type="checkbox"/> A petition has been filed for this unsigned inventor | | | |
| Given Name (first and middle (if any)) | | Family Name or Surname | | | |
| Kevin L. | | Gering | | | |
| Inventor's Signature |  | | | Date | 11/07/00 |
| Residence: City | Idaho Falls | State | ID | Country | US |
| Post Office Address | 2480 Princess Drive | | | | |
| Post Office Address | | | | | |
| City | Idaho Falls | State | ID | ZIP | 83406 |
| | | | | Country | US |

☒ Additional inventors are being named on the 1 supplemental Additional Inventor(s) sheet(s) PTO/SB/02A attached hereto